

ABSTRACTS FOR POSTER SESSION A
2:10-3:40 PM, Thursday, June 6

[A1]

Method for Surface Calculations in a Semi-Infinite Geometry

by Yonas B. Abraham and N. A. W. Holzwarth
Wake Forest University

We are developing a "semi-infinite projector augmented wave (SI-PAW) " method for solving the Kohn-Sham equations in a semi-infinite geometry appropriate for analyzing our photoelectron spectroscopy measurements. This method is especially useful for distinguishing between bulk, surface, and defect states as well as for calculating work function changes.

In this method, we divide the material into three regions. In region I (bulk region), the wave functions are composed of linear combinations of Bloch wave functions of the converged selfconsistent periodic lattice. In region II (intermediate region) the wave functions are composed of extensions of the Bloch wave functions plus evanescent waves which decay into the material. In region III (vacuum region) the wave functions are composed of functions which decay or propagate into the vacuum. The complete wave functions are determined by matching the three forms at the boundaries.

This work was supported by NSF grant DMR-9706575 and a SUR grant from IBM.

[A2]

Full-Potential Local-Orbital Minimum-Basis Scheme (FPLO)

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A high precision total energy code (1mHartree per atom accuracy) is presented which for large unit cells (50 atoms) is about two orders of magnitude faster than FPLAPW. Full relativistic versions, a highly flexible CPA for substitutional disorder, and rotational invariant LSDA+U implementations exist.

[A3]

Vibrational Properties Of Random alloys: a formalism to treat off-diagonal disorder

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A substitutional defect at a site in a crystal produces three types of disorder. In the electronic tight-binding model the diagonal perturbations correspond to changes in the energy level at the defect site, the off-diagonal perturbations changes the interatomic hopping to and from the defect site and the environmental disorder brings in changes in the vicinity of the defect due to effects such as charge transfer and lattice relaxation. In the case of phonon, spin-wave or other Goldstone systems, the treatment of environmental disorder is a necessity because there, it is coupled with other types of disorder by translational or rotational symmetry of the system. The most successful general analytic theory for disordered systems has been the single-site coherent-potential approximation (CPA). Due to its single-site nature, it fails to include the effects of force-constant disorder in the case of phonons in random alloys. Various generalizations of the single-site CPA work only for certain limiting cases or are non-analytic. Here, we propose a generalized formalism for real three-dimensional systems capable of tackling all three kinds of disorder and producing reliable results for phonon properties. We will present results for phonons in random $\text{Ni}_{50}\text{Pt}_{50}$ and $\text{Ni}_{55}\text{Pd}_{45}$ alloys which have large force-constant disorder. We will present calculations on dispersion curves, spectral densities and densities of states and compare them with those of the single-site CPA and with experiment.

[A4]

All electron DFT calculations using a wavelet-like basis

Ivan P. Daykov (Cornell University),
Torkel D. Engeness (MIT),
Tomás A. Arias (Cornell University)

We present the first all-electron density-functional calculations in a solid using a wavelet like basis. We show that this approach is unique in providing systematic “transparent” convergence (convergence with a priori prediction of errors) to tolerances beyond chemical (millihartree) accuracy. This method is thus ideal for exploration of materials subject to novel conditions, where there is little experience with how traditional methods perform, and also for the development and eventual use of chemically-accurate density functionals, where reliable access to millihartree precision is needed.

[A5]

Correlation energy of inhomogeneous systems from RPA-like formalisms

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and Hung M. LE
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Even for weakly to moderately correlated electronic systems, evaluation of the ground-

state energy is by no means a solved problem in practice. Many-body wavefunction methods (DMC, CI, Moeller-Plesset) are unworkable for large complex systems, and the popular Density Functional approaches do not give a reliable treatment of dispersion forces or other cases involving long-ranged correlation holes. Cases in point are polymer cohesion, protein conformation and, probably, graphitic energy storage systems. The inhomogeneous Random Phase Approximation (RPA), unlike its familiar homogeneous-gas counterpart, is not trivial to evaluate numerically but has recently been calculated in simple cases [1,2]. It provides both a simple approximation for the electronic response and, via the fluctuation dissipation theorem, a nontrivial correlation energy that can be shown to contain covalent bonding and to give qualitatively correct van der Waals behaviour. To obtain quantitatively correct van der Waals behaviour, an additional local field correction is required. We discuss some forms for this correction including modified local approximations [3] and a generalisation [4] of the Singwi-Tosi-Land-Sj=F6lander [5] homogeneous electron gas formalism. Benchmarks against QMC are encouraging. =20

[1] J.M.Pitarke and A. Eguiluz, Phys. Rev. B 57, 6329 (1998)

[2] J. F. Dobson and J. Wang, Phys. Rev. Lett. 82, 2123 (1999)

[3] J. F. Dobson and J. Wang, Phys. Rev. B 62, 10038 (2000)

[4] J. F. Dobson, J.Wang and T. Gould, cond-mat/0111447

[5] K. S. Singwi, M. P. Tosi, R. H. Land and A. Sj=F6lander, Phys. Rev. 176, 589 (1968)

[A6]

OHMMS: a framework for multi-scale materials simulations

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John W. Wilkins, Department of Physics, Ohio State University

With computational materials science rapidly evolving and growing in importances, it is critical to develop algorithms in conjunction with new theoretical developments and to accelerate their implementations on high-performance computers of today and in future. The main goal of OHMMS is to provide Object-oriented High-performance solutions for Multi-scale Materials Simulations to meet such challenges. OHMMS achieved i) high flexibility and re-usability by applying object-oriented programming practice, such as design patterns and ii) performance by utilizing advanced abstractions like expression templates for low-level components. Presented are the overview of OHMMS designs for classical and quantum atomistic simulations and the performance of core routines for OHMMS applications on several architectures.

[A7]

Finite-Element Method for Large-Scale Ab Initio Electronic-Structure Calculations

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The finite-element (FE) method is a widely used and quite general approach for the solution of partial differential equations. We discuss its adaptation and application to the ab initio electronic-structure problem in a density-functional context. Like the planewave (PW) method, the FE method is a systematically improvable, variational expansion approach. Unlike the PW method, however, its basis functions are strictly local in real space; and it is from this that the scalability of the method derives. We discuss applications of the method to non-self-consistent ab initio positron distribution and lifetime calculations for systems of over 5000 atoms, and our initial self-consistent results for a variety of test problems.

[A8]

Relativistic NMR Chemical Shifts of Heavy Nuclei with Pseudopotentials

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Francesco Mauri
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Nuclear Magnetic Resonance (NMR) experiments on heavy nuclei present both experimental and theoretical challenges. As measurements of the chemical shift of nuclei beyond the second period are becoming increasingly common, it is important that accurate theoretical techniques are available to predict and interpret these experimental results. First principles Density Functional (DFT) calculations have been shown to provide an excellent description of the chemical shieldings for compounds containing light elements. However to treat heavy element correctly one must account for the effects of special relativity.

We present a method for the calculation of relativistic NMR chemical shifts using pseudopotentials. The method is based on the gauge including projector augmented wave (GIPAW) approach of Pickard and Mauri [Phys. Rev. B **63** 245101, (2001)].

In this paper we show that the GIPAW approach provides a natural framework for the inclusion of relativistic effects in core properties such as chemical shifts. The relativistic nature of the valence electrons close to the core is of paramount importance and we derive relativistic GIPAW operators that take this into account.

The method allows chemical shifts of large, low symmetry structures containing heavy elements to be calculated efficiently.

[A9]

Localized basis sets in electronic structure theory: Solving a large, sparse generalized eigenvalue problem

K. E. Andersen, J. E. Pask, W. E. Pickett
U.C. Davis

The applicability of first principles electronic structure methods, such as density-functional theory, is limited by the size of the physical system. Topical interest in defects, doping, and nanoscale materials all lead to prohibitively large calculations in terms of both storage and cost. Within these calculations, the dominant computational problem is the need to repeatedly solve a standard or (often) generalized eigenvalue problem, resulting from the discretization of the continuous problem. Recent efforts to treat larger systems have focused on using either localized basis set or finite-difference (no basis set) approaches to this discretization. The common aim is to introduce sparse, structured matrices into the resulting eigenvalue problem, such that the cost of a matrix-vector product is $O(N)$ as the size of the system N gets large. The goal is then to find a computational algorithm to solve the eigenvalue problem that uses as few matrix-vector multiplications and as little storage as possible. Here, we discuss a finite-element approach to density-functional theory. Finite-elements are nonorthogonal basis functions constructed from a linear combination of strictly local, piecewise polynomials. Using finite-elements as the basis provides a natural way to deal with the trade-off between cost and accuracy while still retaining the advantages of a basis set. The structure and sparsity of the resulting generalized eigenvalue problem will be discussed, as will a comparison of algorithms to solve it. These algorithms vary from Krylov subspace and Jacobi-Davidson approaches, to more recent minimization techniques of a generalized Rayleigh quotient using the steepest descent or conjugate gradient methods. A comparison to a recently proposed extension of the conjugate gradient method will be made.

[A10]

Density Functional Theoretical Study of Alkanethiol Self-Assembled Monolayers on Au: Effect of the van der Waals interaction

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We have examined alkanethiol self-assembled monolayers (SAMs) on Au(111) surfaces using density functional theoretical calculations. It turns out that S head groups are located close to the bridge site with S-C bond tilted from the surface normal by 50 degrees. Calculated vibrational spectra and S 2p core level shifts agree quite well with experimental results, strongly confirming our results. Although S-Au bonding can be very well described by our calculations, the present GGA functional cannot describe the van der Waals interactions among alkyl-chains, which is important for molecular packing structures. We introduced empirical correction terms to the GGA functional. The correction term is expressed by C_{ij}/r^6 and coefficients C_{ij} were extracted from

accurate quantum chemical calculations and GGA calculations of n-alkane dimers. The GGA functional with correction terms reproduces the interaction energies among alkyl chains very well. The effect of the correction to the SAMs/Au system will be discussed in detail.

[A11]

Pair-state analysis of the eigenstates of an N-electron system

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The energy of an eigenstate of a N-electron system can be expressed in terms of a sum of pair-state energies (eigenvalues of the two-particle reduced Hamiltonian) weighted by two-particle density matrix elements. This relation suggests that pair-state basis functions should provide a useful tool for analyzing many-electron systems. In order to provide a preliminary test of this idea, pair-state decompositions of exact solutions to some Hubbard model systems are examined as functions of the coupling strength.

[A12]

4000-atom LDA supercell calculations of dilute impurities in III-Vs

P. R. C. Kent, Alex Zunger (NREL)

Lin-Wang Wang (LBNL)

Whereas addition of N to III-Vs, e.g. GaAs, creates localized electron states, addition of P to GaAs or Ga to InAs does not. To investigate this phenomena, particularly in Indium compounds where experimental data is lacking, we have performed 4096 atom LDA calculations using a newly developed algorithm combining “charge patching” with the “folded spectrum method”. This method enables us to obtain selected LDA eigenvalues around the band gap of the 4096 atom system, without solving for all of the occupied states.

[A13]

Ab initio theory of solvation

S.A. Petrosyan, Sohrab Ismail-Beigi, A.A. Rigos, T.A. Arias

Cornell Univ.

This poster introduces a new density functional theorem, along with its implementation and examples, for the ab initio study of systems in the presence of a solvent. The theorem provides formal justification for our earlier work[1,2] to approximate the effects of a solution through a direct functional of the electron density by proving that exact

equilibrium properties of a solvated quantum system can be found by minimizing a universal functional $F[n]$ of only the system charge n , where $F[n]$ contains the familiar Kohn-Sham density functional terms of the solute as well as additional terms to describe the solvent.

The resulting theory provides a natural framework for systematic inclusion of terms beyond the traditional dielectric screening, including cavity surface tension and cavitation volume terms for solutions under external pressure. Being variational, the theory allows straight-forward evaluation of forces through the Hellman-Feynman theorem, thereby significantly reducing both implementational complexity and computational cost.

[1] Sohrab Ismail-Beigi, PhD Thesis, MIT 2000.

[2] cond-mat/0007514 (2000).

[A14]

An Order N real-space method for ab initio quantum transport calculations: application to Carbon nanotube-metal cluster assemblies

Qingzhong Zhao, Marco Buongiorno Nardelli, Jerry Bernholc
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First principles electronic structure calculations are today an very important tool for investigating the physics and chemistry of new nano-structures. Based on an order N real-space local orbital ab initio method and an efficient Green's function technique, we have investigated the quantum transport of carbon nanotube(NT)-metal cluster assemblies. The first principle simulations show that carbon nanotube(NT)-metal cluster assemblies behave as effective chemical sensors. We found that gas absorption onto the metal cluster can drastically change both metallic and semiconducting nanotube-metal cluster's electronic properties. In our quantum transport calculation, a metallic NT-aluminum cluster assembly's electrical resistance is found to increase dramatically after NH₃ absorption, while a semiconducting NT-metal cluster assembly's conductivity is greatly enhanced by the absorbed molecule. The observed behavior can be understood in terms of interactions between the molecular species and the nanotube-cluster system, where successive charge transfers between the components tailor the electronic and transport properties. We will discuss the relation between the electronic response and the mechanism of molecular sensing, and present possible ways to improve the detection of different species.

[A15]

Carbon nanotube-metal cluster assembly as a molecular sensor: an O(N) ab initio quantum transport investigation

Qingzhong Zhao, Marco Buongiorno Nardelli, and J. Bernholc
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Using an $O(N)$ real-space optimized orbital method [1], we have investigated the quantum transport properties of carbon nanotube-metal cluster assemblies. Our results show that adsorption of ammonia on a metal cluster can drastically change the electron transport properties of the system, thus enabling single molecule detection. In particular, NH_3 adsorption on a metallic nanotube dramatically increases the resistance of the nanotube-cluster complex, while adsorption on a semiconducting nanotube results in a large decrease in resistance. The observed behavior can be understood in terms of the interactions between the molecular species and the nanotube-cluster system, where successive charge transfers alter the electronic and transport properties. We will discuss the relation between the electronic response and the mechanism of molecular sensing, and present possible ways to improve the detection of different species.

1. M. Buongiorno Nardelli, J.-L. Fattebert, and J. Bernholc, Phys. Rev. B 64, 245423 (2001).

[A16]

A diagrammatic quantum field approach to localized-electron systems

S. A. Bonev and N. W. Ashcroft
Cornell Univ.

We present a diagrammatic language for the variational evaluation of the energy of systems with localized electrons. It is used to develop a convergent series expansion for the energy in powers of overlap integrals of single-particle orbitals. This method gives intuitive and practical rules for writing down the expansion to arbitrary order of overlap, and can be applied to any spin configuration, and to any dimension. Our approach extends previous work by van Dijk and Vertogen [J. Phys.: Condens. Matter 3, 7763 (1991)], Abarenkov [J. Phys.: Condens. Matter 5, 2341 (1993)], and Mouloupoulos and Ashcroft [Phys. Rev. B 48, 11646 (1993)]. From - Thu May 16 10:49:36 2002

[A17]

Ab Initio Forces in Optically Excited States

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U.C. Berkeley

The first-principles GW-Bethe Salpeter (BSE) formalism allows for calculation of optically excited electronic state properties. To date, the method has been limited to working with fixed ionic configurations and, barring inefficient finite difference methods, one does not know how the structure may be optimized in the excited state. We present a recently developed formalism that allows for calculation of excited state forces within the GW-BSE method. This advance allows for efficient optimization of the excited state geometry and the possibility of molecular dynamics and opens the door to the study of subjects such as photoluminescence or excitonic self-trapping. As

a proof of principle and a test of the method, we presents results on the relaxation of photoexcited carbon dioxide and ammonia.

[A18]

Many-body levels of optically excited and multiply charged InAs nanocrystals modeled by empirical tight binding

Seungwon Lee, Jeongnim Kim, Lars Jonsson, John W. Wilkins (Department of Physics, Ohio State University and NCSA/MCC University of Illinois at Urbana-Champaign), Garnett W. Bryant (NIST), Gerhard Klimeck (JPL, Caltech).

Many-body levels of optically excited and multiply charged InAs nanocrystals are studied with an empirical tight-binding model. Single-particle levels of unstrained spherical InAs nanocrystals are described by an $sp^3d^5s^*$ nearest-neighbor tight-binding model including spin-orbit couplings. For the optically excited InAs nanocrystals, first-order corrections from electron-hole Coulomb and exchange interaction to excitonic levels and the oscillator strengths of the excitonic levels determine several low-lying bright excitonic levels. The origins of the bright excitonic levels are explained in terms of strong "intra"- and "inter"-orbital dipole moments between electron and hole levels. Good agreement with photoluminescence excitation experiments is achieved in the size dependence of the three lowest bright exciton energies for nanocrystals with radius larger than 2 nm. For the multiply charged InAs nanocrystals, polarization of a nanocrystal environment is approximated by modeling the environment with a uniform dielectric medium. This polarization model incorporated into the tight-binding model provides a reasonable description of electron and hole addition energies in scanning tunneling spectroscopy experiments.

[A19] **Acceleration of GW Calculations**

J. Aleksi Soininen, Eric L. Shirley, and J. J. Rehr
NIST

The Levine-Louie model dielectric function and its variants can significantly accelerate self-energy calculations. As noted by Hybertsen and Louie (1988), the speed-up factor can be about four. However, self-energy calculations that include local-field effects in principle still involve a double-sum over reciprocal-lattice vectors. Furthermore, the Levine-Louie model dielectric function, as used by Hybertsen and Louie, does not obey the f-sum rule for off-diagonal matrix elements in its original form. Here, we present an alternative model dielectric function, based on the Levine-Louie work but altered somewhat, which obeys the f-sum rule completely, and which allows for self-energy calculations whose calculation time only involve single-sums or single-loops over reciprocal-lattice vectors. This work is supported by the U.S. Department of

Energy (DOE) Grant DE-FG03-97ER45623 and facilitated by the DOE Computational Materials Science Network (CMSN).

[A20]

Ab Initio calculation of the electronic and optical properties of solid pentacene

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Interest in organic crystals for applications in electronic devices has increased greatly after the discovery of extremely high mobilities in pure samples of crystallized pentacene (C_2H_4), which makes this material a good candidate for organic field-effect transistor [J. H. Schoen, Synth. Metals 122(2001), 157]. We have investigated the electronic properties of this material using state-of-the-art ab initio theories in order to clarify the mechanism for mobility in pentacene (band-like motion, as opposed to incoherent hopping motion). The quasiparticle band structure is calculated within the GW approximation for the electron self energy operator. The excitons and optical spectrum of this material are further investigated using the Bethe-Salpeter formalism.

[A21]

Dielectric function and local-field factor of jellium using the Bethe-Salpeter approximation

Argyrios Tzolakis (Department of Physics, University of Illinois at Urbana-Champaign), Eric L. Shirley (National Institute of Standards and Technology), Richard M. Martin (Department of Physics, University of Illinois at Urbana-Champaign and Lawrence Livermore National Lab)

We calculate the dielectric function (both static and dynamic) and the local-field factor for jellium as a function of q and for different values of r_s , using the Bethe-Salpeter approximation. We assume that the particle and hole interact via a statically screened Coulomb interaction. The polarization diagrams that include this interaction are summed to infinite order, leading to a matrix equation for the inverse of the polarization propagator. Employing a string of manipulations we convert the matrices into one-dimensional arrays and we invert the resulting one-dimensional arrays iteratively. This allows us to use matrices of very large size, something that would have been forbidden with “straightforward” inversion, and therefore achieve high levels of accuracy with reduced computational time. Finally, we investigate the role of the forward- and backward-going pairs. For the calculation of the local-field factor we find that the

inclusion of both kinds of pairs gives qualitatively different results compared to the case when only forward-going pairs are included. Our calculations are in qualitative agreement with recent calculations and experiments (S. Moroni, D. M. Ceperley, and G. Senatore, Phys. Rev. Lett. 75, 689 (1995), B. C. Larson et al., Phys. Rev. Lett. 77, 1346 (1996)).

[A22]

All-Electron, Conserving GW calculation of the Quasi-Particle Band Gap in Si and Ge: Effects of the Deep Core States and Many-Body Self-consistency

Wei Ku*, and A. G. Eguiluz (Department of Physics, University of Tennessee)

Surprising results were found in a study of the quasi-particle (QP) band gap in Si and Ge via a novel all-electron, conserving implementation of the GW approximation. The deep core states are shown to play a key role via the exchange diagram, whose treatment via the LDA introduces non-negligible inaccuracy in the calculated gap. On the other hand, the 3d semi-core states of Ge are shown to play no role in the size of the QP gap within the GW approximation, either via their polarization or via their exchange with the valence/conduction states. In contrast to earlier studies, many-body self-consistency is crucial in our all-electron calculation to bring about a reasonable band gap, as a proper implementation of the non-self-consistent all-electron GW approximation results in gaps that are only slightly larger than LDA ones. A detailed analysis of the striking differences between current pseudopotential results and the newly emerging all-electron ones will be given, as well as the large impact of many-body self-consistency on the QP lifetimes.

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Ref: Wei Ku, and A. G. Eguiluz, cond-mat/0203523.

[A23]

Electronic Structure of Carbon Nano-peapods

Youngmi Cho and Jisoon Ihm

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We have performed first-principles pseudopotential calculations to investigate the electronic structure of the fullerenes encapsulated inside the carbon nanotubes, the so-called ‘carbon nano-peapods’. We find that the inserted fullerene molecules modify the local electronic structure of the host nanotube around the Fermi level. The maximum calculated strain in the relaxed geometry is about 1% usually expected. The nearly free electron state of the carbon nanotube is mixed with the fullerene states and the charge distribution is substantially modified. In order to study the behavior of the nearly free electron state under different environments, we introduce a simplified model potential to mimic the carbon nano-peapod system. Comparison with first-principle calculations

shows that the nearly free electron state does not depend on the exact atomic configuration. We also find that the metal atom(s) of the metallofullerene and the metal substrate have a noticeable influence on the electronic structure of the nanotube. The interpretation of the recent STM image of the carbon nano-peapod system is presented.

ABSTRACTS FOR POSTER SESSION B
2:10-3:40 PM, Friday, June 7

[B1]

Itinerant Ferromagnetism and Quantum Criticality in Sc_3In

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The electronic structure and magnetic properties of hexagonal Sc_3In are calculated within density functional theory. We find that the Fermi energy lies in a region of flat Sc d derived bands leading to a peak in the density of states and Stoner ferromagnetism. The calculated local spin density and generalized gradient approximation spin magnetizations are both enhanced with respect to experiment, which is an indication of significant quantum critical fluctuations, neglected in these approximations. We find, as expected, that the ferromagnetism is initially enhanced under pressure, meaning that the critical point cannot be reached with modest pressure. However, we find that the density of states peak around the Fermi energy and the calculated density functional magnetic properties are sensitive to the c/a ratio, so that the quantum critical point may be reached under uniaxial strain.

[B2]

Quantum Monte Carlo treatment of Spin Orbit Interaction

Raghu Chatanathody, Kevin Schmidt, and John Shumway

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We treat the spin-orbit interaction in many-electron systems within variational and diffusion Monte Carlo. We explicitly include the spin degrees of freedom by putting a two-component spinor on each electron. In our all-electron atomic calculations, the spin-orbit interaction is a parameter-free relativistic correction to the Schroedinger equation. This interaction is simply the local electric field at each electron, which appears as a magnetic field in that electron's rest frame. Thus our many-body calculations give the atomic spin-orbit coupling strengths by evaluating the electric field from the nucleus and all other electrons, and coupling to the electron's spin and velocity. We illustrate this method on first and second row atoms, and discuss its potential applications to solids and molecules. (Work supported by DARPA/ONR AWJ9230)

[B3]

Structures and dynamics of adsorbed Si atom on Si(111)-(7×7) surface

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The structures and dynamics of adsorbed Si atom on Si(111)-(7×7) surface are studied by *ab initio* density functional calculations. The potential energy surface (PES) for Si/Si(111)-(7×7) is obtained by mapping out the total energy as a function of its positions on the surface, and a complete adsorption and diffusion picture is established. Inside the faulted and unfaulted regions, many stable adsorption sites are found near the adatoms and the rest atoms because strong dangling bonds exist on top of these atoms. We also find stable configurations where the adsorbed Si atom and the adatom form an ad-dimer on top of the T₄ sites (adatom sites). These ad-dimer states are more stable than adsorbed on the dimer row and corner hole regions due to the higher coordination of ad-dimer states to the substrate backbond atoms. As these many stable adsorption sites are so close to each other, when adsorbed Si atom migrates on the Si(111)-(7×7) surface, the breaking and formation of bonds between the adsorbed atom and the surface atoms take place at the same time, and thus led to the surprising low energy barriers; less than 0.3 eV for diffusion surrounding the H₃ site, less than 0.65 eV for diffusion surrounding the rest atom site. For hopping-exchange diffusion along the connecting line of center and corner adatoms through the ad-dimer state, the energy barrier is less than 0.70 eV. For diffusion between faulted and unfaulted halves, by connecting adsorption sites near the dimer row, three kind of optimized diffusion paths are found with energy barriers ranging from 0.96 to 1.21 eV in excellent agreement with experimental results. Due to the existence of dangling bonds on top of the adatoms and rest atoms and the low energy of ad-dimer states, the diffusion inside the faulted and unfaulted regions are much smaller than the diffusion barriers crossing the dimer row regions.

[B4]

Surface stress as the driving force for assembly of Bi nanolines on Si(001)

David Bowler

Department of Physics and Astronomy

University College London

The Bi nanolines which form spontaneously on the Si(001) surface are remarkable for their perfection, straightness and length - nanolines are often more than 400 nm long while being uniformly 1.5nm wide, and a kink in a nanoline has never been observed. The lines are actually quantum anti-wires, but may well have important applications as templates for nanowires. Previously proposed models for the structure of the Bi nanoline have been invalidated by recent scanning tunneling microscopy (STM) data; after an exhaustive search of possible structures, a new model is proposed which agrees

with STM and other experimental observations. This model is remarkable in that it involves reconstruction down to the fifth layer of silicon. The search for the structure of the nanolines elegantly illustrates the benefits of combined experimental and theoretical investigations of nanoscale structures.

[B5]

**Realization of a Large J2 Quasi-2D Spin Half Heisenberg Systems:
Li2VO₂SiO₄ and Li2VO₂GeO₄**

H.Rosner(1), R.R.P.Singh(1), W.H.Zheng(2), J.Oitmaa(2), and W.E.Pickett(1)

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Recent progress in understanding the behavior of unusual magnetic insulators (CaV₄O₉, CaV₃O₇, La₂BaCuO₅) has been achieved only after density functional based calculations were applied to quantify the magnetic state and the hopping amplitude between magnetic ions. Here, applications of LDA methods to the square lattice J1-J2 Heisenberg systems Li₂VO₂GeO₄ and Li₂VO₂SiO₄, the latter showing a phase transition to columnar order at low temperatures, are described. With LDA determined exchange constants, high temperature expansions (HTE) are developed to give the uniform susceptibility and specific heat of the square-lattice J1-J2 Heisenberg models. Combined with a perturbative mean-field theory, we obtain accurate results for the uniform susceptibility of the small-J1/J2 Heisenberg model at all temperatures. Using the HTE results we show that the specific heat and uniform susceptibility of these materials are well described by a small J1/J2 Heisenberg model in agreement with the LDA predictions. Furthermore, the measured Neel temperature is consistent with our LDA derived J_c exchange constants. We suggest that the observed small moments in the NMR spectra are due to a large cancellation coming from the 3-dimensional geometrical structure and not from a proximity to a critical point. Further experiments which would be particularly suited to an accurate determination of the J1/J2 ratio for these systems are discussed.

[B6]

Simulation of Defects in Silicon

Kaden R.A. Hazzard

Ohio State Univ.

Central to technology development is the ability to rapidly develop new materials. Neither industry nor government has the time or money to fund 10-15 years developing new materials for a specific application. In the case of semiconductors, a principal challenge is reducing the feature size on the computer chip. One identified obstacle is that implanted dopants diffuse faster – by a factor of 10 to 100 – than they do

when the material is thermally annealed to remove the defects. The belief is that implantation produces interstitial atoms (atoms not at a site of the perfect lattice) and these interstitials somehow (the mechanism is still not verified) speed the diffusion of the dopants.

This study is aimed at understanding the diffusion of small numbers of interstitials. We know that these interstitials form extended defects, which provide sources of interstitials which can aid the dopant diffusion. But how do these large interstitial defects grow? To gain insight into this, we have studied the motion of small silicon interstitials clusters – clusters of two interstitials (di-interstitials) and clusters of three (tri-interstitials).

We use molecular dynamics to study diffusion. We compute the force on a di-interstitial (say) and move it under the force for a short time – typically 2 femtoseconds. Then we re-compute the force and iterate. At temperatures from 800 K to 1100 K we accumulate over a hundred million time steps (almost a microsecond), an unprecedented calculation. Of course, no one (not even a junior) would look at a million frames trying to see if anything happened, especially since most of the time the atoms just vibrate. Fortunately, the group has developed an approach that can look for large motions over time scales varying from 4 femtoseconds to 8 picoseconds. By saving only the long-term stable structures we reduce the number of "frames" to a few thousand. Another undergraduate colleague, Steve Barr, and I looked at each frame individually. To my boss's surprise, we find several low-lying structures - that is, they are accessible in the relevant temperature range for annealing and device application. I didn't realize how surprising it was until I looked in the literature found no reference to them. These thermally accessible structures are interesting for two reasons: (1) They offer multiple routes for the rapid diffusion of interstitial defects. Indeed, the tri-interstitials move more rapidly than the di-interstitials that move, in turn, more rapidly than single interstitials. (2) Some of the thermally accessible defects are elongated – so much so that we speculate they may be the precursors for forming extended structures.

In the future we want to look at larger interstitial clusters and develop less time intensive methods of identifying and cataloging the structures. For a 2nd year student it is very exciting to be at the beginning of what my boss calls a "new era in the growth and nucleation of defects in solids" – these defects determining the industrially useful properties of the materials – as we strive to more quickly develop new materials.

[B7]

Path Integral Simulations of Si/Ge Quantum Dots

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We have developed a quantum Monte Carlo technique for simulating the electronic and optical properties of self-assembled quantum dots, starting from an experimentally observed structure. We derive an effective mass hamiltonian from a structural model by solving for strain fields and computing effective confinement potentials. We then apply path integral Monte Carlo directly to this effective mass hamiltonian, so that all

interaction effects (mean-field, exchange, and correlation) are solved directly, bypassing the calculation of non-interacting single-particle states. The path integrals allow us to calculate exciton, charging, and multiexciton energies, as well as a variety of other physical properties, such as dipole moment, charge densities, and radiative lifetimes. We show our initial results on Si/Ge dots, and discuss extensions to other nanosystems and multiband effective mass models.

[B8]

Ab Initio investigations of the Structural Properties of Hexagonal Graphite under High Pressure

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The structure of hexagonal graphite under high pressure has been a puzzle to experimentalists for some time, though several structural measurements such as high pressure X-ray diffractions [1] and high pressure Raman scatterings [2] have been reported. Some people [1] believe that the structure is a diamond like structure of sp³ bonding based on their x-ray experiments while the others [2] suggested that the structure a layered structure of sp² bonding based on their Raman scattering experiments. To figure out this contradiction, we have performed first principles ultrasoft pseudopotentials calculations to determine the high pressure structure of hexagonal graphite. We used Vienna ab initio simulation package. We then used the resultant high pressure structure to simulate the experimental X-ray diffraction pattern. We have also analysed the electron charge density distribution to see the bonding character of the the structure. Based on these results, we could conclude that the hexagonal graphite under hydrostatic pressure should be a layered structure of sp² bonding. Further theoretical calculations of the phonon dispersion relations are in progress.

[1] T. Yagi, et al., Phys. Rev. B 46 (1992) 6031.

[2] T. L. Schindler, et al., J. Phys.: Condens. Matter 33 (1995) L637.

[B9]

Electronic Structure Analysis of FeI₂

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The compound FeI₂ shows a rather unusual ordered spin structure at low temperature, although the isostructural FeX₂ halides (X=Cl, Br) order antiferromagnetically in a simple way. The application of magnetic fields and pressure reveals a very rich phase

diagram showing antiferromagnetic, ferrimagnetic and several other magnetic phases as well as a transition from insulating to metallic behavior. In order to investigate the electronic and magnetic structure in preparation for studying the exchange couplings between Fe-ions, we have applied several full potential and non-full potential band structure methods, along with local density approximation and general gradient approximation. For the non spinpolarized case, all calculations result in an insulator with a very small band gap (0.2 eV for LMTO (LDA), 50 meV for LAPW and FPLO (LDA), and 250 meV for LAPW (GGA). Our spin polarized calculations give various FM and AFM solutions, all of them describing half metallic behavior and showing very similar total energies. The calculated metallic behavior is in contradiction with the experimentally observed insulating ground state. This discrepancy is probably due to the strong correlations in the Fe-3d states. Work in progress using LDA+U is expected to resolve this discrepancy

[B10]

Magnetic Ground State Calculations in Manganite Antiperovskites GaCMn₃ and CuNMn₃

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In light of the recent discovery of superconductivity near a ferromagnetic instability in MgCNi₃, it becomes important to investigate isostructural compounds built on 3d elements. GaCMn₃ and CuNMn₃ are both metallic antiperovskite structures with magnetic transitions. CuNMn₃ undergoes a lattice transition from cubic to tetragonal as well as a magnetic transition from paramagnetic to ferrimagnetic (at T=150K). GaCMn₃ remains cubic across its magnetic phase change from ferromagnetic (at T=255K) to antiferromagnetic (at T=160K). The ground state electronic and magnetic properties of these two compounds are calculated using two different full-potential basis sets to compare results. The magnetic ground state of both compounds is correctly reproduced and the nesting properties of the fermi surface are reinvestigated and found to be in contrast with previously published non-full-potential results. The magnetic moment and bandstructure are independent of basis set, as expected, but are strongly dependent on the choice of exchange-correlation (XC) potential. These compounds provide a striking example that LDA and GGA do not produce similar results in some magnetic systems, though non-magnetic calculations are identical. Fixed spin moment calculations are carried out in order to investigate the discrepancy between results obtained with different XC potentials.

[B11]

Path integral Monte Carlo calculation of radiative recombination rates in quantum dots

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Confinement, correlation, and mean-field interactions compete to determine radiative recombination rates in quantum dots. Recent time-resolve photoluminescence experiments find that the biexcitons decay about 1.5 times faster than single excitons. This places the system somewhere between the strongly confined and bulk limit. Previous theoretical approaches to this problem using the wavefunction representations of quantum mechanics have been unable to quantitatively solve a realistic model. We develop a Feynman path-integral formulation of the problem that allows the direct evaluation of the recombination rate, including thermal and many-body effects. We evaluate the path-integral expression using real-space Monte Carlo integration. We apply this method to several different dots, using a model that includes an experimentally motivated lens-shaped confining potential and anisotropic effective masses, and compare to recent experiments.

[B12]

Instabilities in the Cis-Trans Isomerization, and Singlet-Triplet Gaps in Push-Pull Compounds

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The energy barriers for the cis-trans isomerization are obtained for a sample of push-pull compounds. The appropriate trend in the values for the barriers is obtained when the instability of the Restricted Kohn Sham (RKS) solution with respect to the Unrestricted one is taken into account. By using a non-symmetrical push-pull compound, the width of the instability region is determined as 50 degrees around the transition state. The results in this system show that, the instabilities in the twisted configurations of double bonds are related mainly to spin symmetry effects rather than to the symmetry of the external potential. The description of the singlet-triplet gap along the rotation by the use of spin potential and hardness is qualitatively preserved but on a quantitative basis it only remains in the region free of instabilities.

[B13]

Electron-Phonon Interactions in Field-Effect Doped Layers of Crystalline C_{60}^*

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Recent developments [1] have shown that MOS-FET devices can induce 2-D metallic layers in C_{60} with a high carrier density. For 2-4 holes per C_{60} , superconductivity has been observed with transition temperatures T_c as high as 52 K in pure C_{60} [1a] and 117 K in intercalated C_{60} [1b], the highest T_c reported for any non-copper-oxide superconductor. Density functional calculations [2] have shown that the carriers are confined to 2-D bands, with states that are highly distorted due to the field and 2-D Fermi surfaces that are very sensitive to the orientational order. In this work we study the electron-phonon interactions for the metallic interface states, motivated by the evidence that superconductivity in the bulk is electron-phonon mediated. We report our initial results for the behavior of the electron-phonon coupling under field-effect doping. Tight binding and DFT calculations (using the SIESTA code [3]) are performed for a range of electron and hole densities in various ordered C_{60} layers.

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[1] [a] J. H. Schön, Ch. Kloc and B. Batlogg, *Nature* 408, 549 (2000); [b] *Science* 293, 2432 (2001).

[2] J. L. Mozos, P. Ordejón and R. Martin, to be published.

[3] P. Ordejón E. Artacho, and J. M. Soler, *Phys. Rev. B* 53, R10441 (1996).

[B14]

The Mechanism of the Titanium HCP to Omega Transformation

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Under pressure, titanium transforms from a hexagonal-closed-packed (hcp) structure to a high pressure omega phase. The phase transition from hcp to omega is known to be martensitic, but the transformation mechanics have not been described quantitatively. To find the most probable mechanism, we produced a set of possible mechanisms by enumerating common subgroups of the two structures. The list was reduced by keeping only mechanisms with the smallest strains. An estimated energy barrier for each remaining mechanism was calculated using a tight-binding model for titanium. From this set of approximate barriers, the mechanisms with the smallest barriers were chosen, and the true energy barrier was calculated using an ab initio method. Calculating the barrier as a function of pressure both indicates the transformation mechanism and permits an estimate of the zero temperature transformation pressure.

[B15]

Role of Impurities in the Titanium hcp to omega Transformation

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Titanium, like all group IVb transition metals, shows martensitic phase transitions between its bcc, hcp and omega phases. For temperatures below about 1000 K hcp-Ti is stable at low pressure and undergoes a martensitic phase transformation to the omega phase at about 8 GPa. Experiments indicate that impurities strongly influence this transition: While small amounts of V favour the omega phase, O impurities with concentrations as low as 3000 ppm shut down or retard the transition. Ab initio nudged elastic band (NEB) calculations show that the Burgers [111] mechanism has the lowest energy barrier of 9 meV/atom for this transition. For this pathway NEB calculations determine the change in the energy barrier due to substitutional V and Al impurities as well as interstitial C, N, and O atoms of about 2 concentration. The impurities change the energy difference between the hcp and omega phase by less than 3 meV/atom. Substitutional impurities only weakly influences the energy barrier. The interstitial impurities on the other hand increases the energy barrier by factor of two, impeding the hcp to omega transformation.

[B16]

Finite Size Scaling for Quantum Critical Phenomena

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The finite size scaling ansatz is combined with the variational method to extract information about critical behavior of quantum Hamiltonians. This approach is based on taking the number of elements in a complete basis set as the size of the system. As in statistical mechanics, the finite size scaling can then be used directly to the Schrodinger equation. This approach is general and gives very accurate results for the critical parameters, for which the bound state energy becomes absorbed or degenerate with a continuum. To illustrate the applications in quantum calculations, we present detailed calculations for stability of atomic and molecular systems and size effects in the electronic properties of quantum dots.

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*Phys. Rev. E 64, 056120-1-6 (2001).

[B17]

Ab initio calculation of charged divacancy in silicon

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We report what is to our knowledge the first ab initio study of the singly charged

divacancy V_2^- in silicon to make direct comparison with mechanical response experiments. This novel approach does not rely upon energy differences, which have proven to be too small (0.1 eV) to resolve reliably within the uncertainties of the LSDA, but rather on a calculation of the full activation volume tensor of the defect, which is independent of energy differences. This allows us to make a definitive determination of the ground-state structure of the defect and thereby resolve an on-going theoretical debate. We present results from plane-wave, pseudopotential calculations within the local spin-density approximation (LSDA) for the activation volume tensor of competing forms for the structure of the defect and then compare these theoretical results to the pioneering EPR experiments of Watkins and Corbett(1). As an application, we present a new way of determining the quality factor from the activation volume tensor.

(1) Watkins, G. D. and Corbett, J. W. , Phys Rev, 138, A543 (1965)

[B18]

Socorro: A modular extensible framework for electronic-structure calculations

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Electronic-structure calculations based on density-functional theory (DFT) are being utilized more and more to determine and understand the behavior of complex materials. While the theoretical foundations underlying DFT and related techniques are quite well developed, computer codes that implement these techniques generally do not conform to modern software engineering practices. This makes them difficult to modify, thereby impeding the implementation and use of new electronic-structure techniques and decreasing the productivity of researchers. Socorro is our effort to develop a flexible, easily maintained, open-source electronic-structure code. This code is mainly written in Fortran 90/95 + MPI, and widespread use has been made of object-oriented features of Fortran 90/95 such as modules, derived types, data-hiding, and operator overloading. A Socorro module encapsulates a data structure, whose details are kept private, along with public routines used by code outside the module to manipulate the data structure. Since outside code does not depend on details of the module's data structure, the implementation can be changed independently of the outside code. The flexibility inherent in this approach is greatly enhanced by the use of a specification-driven methodology where the focus is on careful design of interfaces between modules. Socorro is organized as a hierarchy of modules where upper level modules group and enforce constraints between objects defined within lower level modules. Details of this structure and how it facilitates extensibility will be discussed. Considerable effort has also been devoted to maintaining high performance in an object-oriented framework. The tools that we have found useful to achieve this include lazy copying, data polymorphism, and light-weight comparisons. Socorro is available (under a GNU public license) for use as a platform on which to develop new electronic-structure techniques and as

a high-performance tool for studying the behavior of complex materials. The current version of Socorro implements the local-density approximation (Ceperley-Alder) to exchange and correlation, three forms of the generalized-gradient approximation (PW91, PBE, and BLYP), molecular dynamics, the dimer method for finding transition states, and the projector-augmented wave technique.

[B19]

Wannier State Analysis of insulating ferromagnetism in $\text{La}_4\text{Ba}_2\text{Cu}_2\text{O}_{10}$

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The exceptional insulating ferromagnetism, discovered in the brown phase of half-filled $\text{La}_4\text{Ba}_2\text{Cu}_2\text{O}_{10}$ ($\text{La}422$), poses a great challenge in our quantitative theoretical understanding of quantum magnetism. It is even more puzzling with the recent discovery of the "normal" antiferromagnetism of the isostructural $\text{Nd}_4\text{Ba}_2\text{Cu}_2\text{O}_{10}$ ($\text{Nd}422$). With a newly developed scheme, in which the reformulated ab initio 2nd quantized Hamiltonian is represented based on energy-resolved all-electron Wannier states (WSs), the origin of the ferromagnetism is identified to be the intersite direct exchange between WSs. The calculated parameters (t , U , and J) indicate that this mechanism overwhelms the Hubbard-type superexchange considered in existing analysis. By contrast, $\text{Nd}422$ is shown to develop the experimentally observed antiferromagnetism via its characteristics of a 1D chain. Contrary to current assumption, in both compounds, the dominant in-plane coupling turns out to be not with the nearest neighbors, but with the ones above/below them. These results agree very well with many aspects of current experimental observations. The energy-resolved WSs are shown to be ideal for studying magnetism from first principles, and the spatial distribution of the WSs reflects that of the spin moment. The crucial role of the chemical replacement is analyzed with additional calculation on $\text{La}422$ under pressure, which not only reproduces observed magnetic order, but also suggests an intriguing pressure-induced ferromagnetic to antiferromagnetic transition upon higher pressure.

Ref: Wei Ku, H. Rosner, W. E. Pickett, and R. T. Scalettar, cond-mat/0205300.

[B20]

Chemistry of sulfur oxides on Pt surfaces

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We have applied first-principles density functional theory computational methods to the sulfur poisoning problem in automotive catalysis. The thermodynamics of sulfur

oxides, including O, S, SO, SO₂, SO₃ and SO₄, were explored exhaustively on Pt(111) and related stepped surfaces. The reaction path of O + SO₂ → SO₃ and the diffusion paths for both O and SO₂ were computed on the Pt(111) surface. We found that adsorbate interactions could be modeled as repulsive dipole-dipole interactions.

[B21]

Ab initio studies of structural stability and magnetism in Ni₃In

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To understand the structural stability and magnetism in the intermetallic compound Ni₃In, we have performed total energy and electronic structure calculations for Ni₃In as well as Ni₃Al in the cubic L1₂, tetragonal D0₂₂ and hexagonal D0₁₉ structures. The highly accurate full-potential linearized augmented-plane-wave method has been used. The calculations are based on first-principle density functional theory with generalized gradient approximation. The theoretical equilibrium lattice constants and bulk moduli of both the compounds are in good agreement with available experiments. Surprisingly, unlike Ni₃Al and other related intermetallics, Ni₃In in the nonmagnetic state is predicted to energetically favor the D0₂₂ structure rather than the L1₂ structure. However, the L1₂ and D0₁₉ structures are found to be magnetically unstable while the D0₂₂ structure is not. Ferromagnetism would make the L1₂ structure slightly lower in energy than the D0₂₂ structure by merely 7 meV/formula-unit. Therefore, the present theoretical work suggests that Ni₃In at low temperatures is a weak ferromagnet and undergoes a structural and magnetic phase transformation as the temperature is raised to the room temperature. This picture would allow one to consistently interpret previous structural experiments and measured x-ray absorption spectra. Moreover, it is also predicted that unlike Ni₃Al and other related intermetallics, there would be a pressure-induced structural phase transition from the L1₂ to D0₂₂ in Ni₃In at low temperatures. It is hoped that these interesting findings would stimulate further experimental investigations such as temperature-dependent structural, specific-heat and magnetization experiment on this nearly or weakly magnetic intermetallic compound..

[B22]

First-principles design of light-emitting silicon

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Indirect-gap crystalline Si emits neither efficiently, nor in the visible range. To add

optical functionality, various approaches have been proposed: e. g., the epitaxial growth of light-emitting III-V materials or novel group-IV based materials. Despite some successes in these approaches, they either endure a large lattice mismatch, growth difficulty, or their synthesis can be extremely difficult. Here, we explore a new direction in modifying Si band structure by impurities. For example, after smartly adding oxygen atoms, (Si)₈/O (001) superlattice structure exhibits 1.5-eV direct gap with significantly enhanced optical transition matrix element. We find that the oxygen atoms introduce new non-tetrahedral bonds and extra electronic states. These new states interact with the Si host states and induce changes in the energy levels and host wavefunctions. Obviously, this is a chemical effect, not quantum confinement. We show that such band gap engineering can be fully understood and manipulated. This opens new ways to control Si properties as desired.